



A preliminary investigation into the physical and chemical properties of biomass ashes used as aggregate fillers for bituminous mixtures

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ARTICLE INFO

Article history:

Received 8 September 2012

Accepted 17 May 2013

Available online 19 June 2013

Keywords:

Biomass ash

Waste management

Filler

Bituminous mixtures

Leachability

ABSTRACT

Fly and bottom ashes are the main by-products arising from the combustion of solid biomass. Since the production of energy from this source is increasing, the processing and disposal of the resulting ashes has become an environmental and economic issue. Such ashes are of interest as a construction material because they are composed of very fine particles similar to fillers normally employed in bituminous and cementitious mixtures. This research investigates the potential use of ash from biomass as filler in bituminous mixtures. The morphological, physical and chemical characteristics of 21 different ashes and two traditional fillers (calcium carbonate and “recovered” plant filler) were evaluated and discussed. Leaching tests, performed in order to quantify the release of pollutants, revealed that five ashes do not comply with the Italian environmental re-use limits. Experimental results show a wide range of values for almost all the investigated properties and a low correlation with biomass type in terms of origin and chemical composition. Furthermore, sieving and milling processes were found to improve the properties of the raw material in terms of grading and sample porosity. The effectiveness of these treatments and the low content of organic matter and harmful fines suggest that most of the biomass ashes investigated may be regarded as potential replacements for natural filler in bituminous mixtures.

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1. Introduction

European Directive 2009/28/CE defines biomass as the biodegradable fraction of products, waste and residues from biological origin from agriculture (including vegetal and animal substances), forestry and related industries including fisheries and aquaculture, as well as the biodegradable fraction of industrial and municipal waste. It is essentially any organic matter which contains stored solar energy and can, therefore, be used as an energy source. In this context, fossil fuels are not considered to be biomass.

The production of energy deriving from biomass combustion and consequent generation of its by-products is increasing worldwide: in Italy, in 2009, the gross domestic consumption from solid biomass combustion was 4.1 Mtoe, with this figure being a 5% and a 75% increase on the consumption figures for 2008 and 2000 respectively (ENEA, 2009).

Fly and bottom ashes represent the main residues of a combustion process: they are composed of mineral materials (in their oxidised form) absorbed by the biomass during its lifecycle or incorporated during harvesting and of a small quantity (up to

20%) of unburned organic matter. The amount of ashes produced depends on combustion chamber conditions and biomass type, with values that vary between 2% (i.e. woodchips) and 20% (i.e. rice husk). Bottom ashes, which settle under the grate of the combustion chamber, are the coarsest and heaviest constituent part of the combustion by-products, while fly ashes, which remain suspended in the flue gases, are the finest part. Fly ashes represent up to 40% of the total ashes produced and are usually removed through electrostatic precipitators and fabric-filter baghouses.

The recycling of biomass ashes as construction materials meets the recommendations of the [European Directive on waste 2008/98/CE](#) and has significant environmental benefits related to the decrease in the quantity of natural aggregates extracted from quarries and to the reduction of waste carried to landfills. In Italy, since the European Waste Catalogue ([European Commission Decision 2000/532/CE](#)) classifies waste from biomass combustion as a non-hazardous material, a simplified procedure ([Regulation DM 05/02/98](#)), requesting a communication of intent rather than a submission for formal authorisation for recycling these types of ashes, is sufficient. According to this regulation, biomass ashes can only be recycled in concrete, cement and brick production, in embankment construction and environmental re-use, but not in other applications such as the production of bituminous mixtures. In this case, the usual authorisation procedure is required. In Italy, as a result of legal constraints, the majority of these ashes are taken

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to landfills, involving considerable transportation, processing and disposal costs. Their re-use in bituminous materials could make a significant contribution to a reduction in ash disposal costs when one considers that 34.9 million tons of bituminous mixtures were produced in Italy in 2009 (EAPA, 2009).

Bituminous mixtures are basically composed of aggregates of different sizes, filler and bitumen. Fillers are mineral grains most of which pass at 63 μm sieve (EN 13043), and represent 5–10% of the aggregates by weight in the whole mixture. Filler can have a natural origin when derived from the crushing of rocks, or can be manufactured in industry as in the case of lime, cement, ash and slag. Although its main function is filling the voids in the aggregate skeleton to create a denser mixture, several studies have demonstrated that filler has other important roles. Depending on its particle size and structure, it stiffens and/or extends the binder (Kandhal and Parker, 1998; Grabowski and Wilanowicz, 2008), consequently affecting the occurrence of rutting and fatigue phenomena. Furthermore, filler also modifies the ageing processes (Gubler et al., 1999; Recasens et al., 2005) and its finest part may act as an anti-stripping agent preventing moisture damage (Kandhal and Parker, 1998).

Although its importance is well recognised, most recent regulations on filler for bituminous mixtures (EN 13043, ASTM D242, AASHTO M17) establish limits for only a few characteristics such as grading, water content, plasticity index and organic content. In addition, the Superpave volumetric mix design system (Cominsky et al., 1994) defines a limit for the quantity of filler in the mixture (corresponding to a filler/binder ratio in the range 0.6–1.2 by weight). The above mentioned characteristics are necessary primarily for quality control, but are not sufficient to obtain information correlated with the expected performance of bituminous mixtures. This is even more evident for manufactured fillers, like biomass ashes, which often exhibit unique behaviours.

The study described in this paper is part of RICCO, a 3-year research project financed by the Italian Ministry of Agricultural, Food and Forestry Policies which aims to investigate the possible re-use of biomass ash as filler in bituminous mixtures. In particular, the goal of the first part of the project is to characterise several ashes from biomass combustion by integrating the tests included in the technical standards on filler for bituminous mixtures EN 13043, with those tests that ongoing research has identified as reliable performance indicators. Furthermore, since it is important to substantiate the hazardous nature of ashes, their environmental impact can be assessed by the European regulation EN 12457-2 on the evaluation of leaching potential.

2. Background

Several studies have been carried out in recent years to assess the possible re-use of biomass ash and its recycling as a substitute for aggregates in concrete mixtures (Martin Morales et al., 2011), in cement production (Ajiwe et al., 2000), and also as a fertilizer (Insam and Knapp, 2011). In road applications, due to its cementing and pozzolanic properties, most of the researches focused on the recycling of biomass ash in soil stabilization (Basha et al., 2005; Nordmark et al., 2011). The recycling of biomass ashes as filler in bituminous mixtures has been studied in The Netherlands: an extensive study led by the Energy Research Centre of The Netherlands (Pels et al., 2006) showed that fly ashes from the gasification of solid biomass are a valuable alternative to natural fillers. Sarabèr and Haasnoot (2012) investigated the physical and chemical characteristics of several fly ashes derived from the combustion of different solid biomasses. Their experimental results indicated that most of the ashes do not meet the requirement for passing 125 and/or 63 μm , concluding that, due to their gradation, biomass

fly ashes can compete more with Municipal Solid Waste Incinerator fly ash and sewage sludge ash than coal fly ash when recycled as filler in bituminous mixtures. Unfortunately, the physical–mechanical properties of ash–bitumen mastics, slurries or mixtures were not investigated. Moreover, the investigation by Sarabèr and Haasnoot (2012) does not take into account the fact that characteristics like gradation, water solubility and soundness are insufficient for an assessment of the use of biomass ashes as filler in bituminous mixtures, as clearly indicated in the present paper.

The majority of the studies concerning the possible re-use of ashes as filler in bituminous mixtures investigate the behaviour of fly and bottom ash derived from coal combustion. A great quantity of ash (20–30% of the original matter) is produced from this combustible which still represents the most widespread energy source in many countries. In the European Union (EU 15) 61 million tons of coal combustion products were produced in 2006 (European Coal Combustion Products Association, 2006), while in the US 125 million tons were produced in 2009 (American Coal Ash Association, 2009). Kavussi and Hicks (1997) evaluated the properties of bituminous mixtures containing four different fillers (limestone, quartz, coal fly ash, kaolin): they found that mastics containing fly ash were more susceptible to brittle failure because of their high porosity, caused by the presence of very small air bubbles formed during the burning process. Sharma et al. (2010) demonstrated that coal fly ash having high calcium content exhibits anti-stripping properties. In order to avoid excessive stiffening of bituminous mixtures, they suggested a maximum value of 60% for the ratio between bulk volume of compacted filler and total filler–bitumen volume. This study also showed that high values for clay content in the ash, as revealed by the Methylene Blue (MB) test, were correlated to low tensile strength ratio and retained stability values, proving that this test can also provide a good estimate of moisture susceptibility.

In recent years the National Cooperative Highway Research Program Project 9-45 (NCHRP, 2010) was developed in order to address the theme of filler role in bituminous mixtures. The study considered 32 different fillers for which the effect on bituminous mixture performance was thoroughly investigated. Out of the total set of analysed fillers, three were fly ashes collected from coal combustion plants. The research identified fractional voids, size distribution, content of calcium compound and active clay content as the most relevant properties for the characterisation of fillers. Compared to mineral fillers routinely used for paving applications, the properties of fly ash encompassed a wider range of values. As a consequence, the prediction model developed to estimate mastic properties from filler characteristics was applicable only to natural fillers. The study concluded that manufactured fillers have a unique influence on mastics and mixtures, with this effect requiring a more detailed investigation.

Although coal ash is the one most studied, many investigations deal with the possible re-use of other ashes in bituminous mixtures. Xue et al. (2009) investigated the effects of Municipal Solid Waste Incinerator (MSWI) ash on Stone Mix Asphalt (SMA). The use of 16% of MSWI ash meets the requirements of Marshall and Superpave mix design procedures. However, the ash lowers the water damage resistance of the mixtures due to its low CaO content causing poor adhesiveness between asphalt and ash. Hassan et al. (2007) studied the effect of replacing 0–3 mm natural aggregates with up to 40% of MSWI ash in bituminous mixtures: the Marshall mix design showed that optimum asphalt content increased significantly as more ash was introduced into the mix, owing to the high absorption properties of the ash.

One solution to the problem of this increasing absorption of binder is represented by vitrification. Bassani et al. (2009) studied a bituminous mixture in which up to 32.5% of 0–2 mm natural

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